CORROSION INVESTIGATIONS OF Al-Si CASTING ALLOYS IN 0.6 M NaCl SOLUTION

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Abstract

This paper presents results of the corrosion investigations of specimens made from finished parts for the automotive industry, produced by high-pressure die casting and gravity die casting process of six Al-Si alloys (40000 series). Open circuit potential and potentiodynamic polarization measurements have been performed using a potentiostat with three-electrode set-up in 0.6 M NaCl naturally aerated solution. Microstructural characterization before and after electrochemical investigations has been carried out with optical microscope to establish the connection between microstructure and corrosion parameters of investigated alloys and to analyze and record surface changes of each sample due to electrochemical corrosion. All alloys show good corrosion resistance, which manifests with low values of corrosion rates, calculated from the corrosion current densities obtained from potentiodynamic polarization measurements. Differences in electrochemical behavior appear due to the distinctions in their chemical composition and microstructure. The type of casting process does not affect electrochemical behavior of Al-Si alloys.

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1 Introduction

The good combination of properties of aluminium based alloys makes these materials increasingly used in a broad spectrum of lightweight structures in transportation and construction. While the automotive industry continues to rely heavily on steel, the need to increase fuel efficiency and reduce CO_2 emissions has led to considerably wider use of aluminium alloys. The average content of aluminium alloys in the car is expected to increase by 60 % by the year 2025 [1, 2].

Aluminium is one of the lightest engineering metals with a density of only 2.7 g cm^{-3}, approximately one-third as much as steel (7.83 g cm^{-3}). Copper, magnesium, manganese, silicon, and zinc are used as the major alloying elements in aluminium alloys [3-5]. The predominant reason for alloying is to increase strength, hardness, and resistance to wear, creep, stress relaxation or fatigue. The influence on these properties is specific to the different alloying

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elements and combinations of them and is related to their alloy phase diagrams and to the microstructures and substructures that they form as a result of solidification, thermomechanical history, heat treatment and/or cold working [4, 6].

High corrosion resistance is another major advantage of aluminium, due to thin, protective oxide film that is bonded strongly to materials surface. This film is stable in aqueous media when the pH is between 4.0 and 8.5. If damaged, film re-forms immediately in most environments [6-8].

Alloying aluminium with other metals to improve its mechanical properties often reduces its corrosion resistance [8-11]. Therefore, the effect of alloying elements on corrosion resistance and mechanical properties has been extensively investigated using various series of aluminium alloys [5, 12-17].

The most significant part of all casting manufactured materials belongs to the aluminium alloys with silicon as the major alloying element. As a result of an excellent combination of castability and mechanical properties, as well as good corrosion resistance and wear resistance, these alloys have a wide range of applications in the automotive and aerospace industries. Al-Si alloys have been used in production of various finished parts for automotive industry, such as turbo-compressor housings, backplates, inserts, turbine housings, central housings and nozzles, as well as exhaust system parts, flywheels, engine and accessory brackets, handles, turbo pipes, brake discs and brake drums. They are mostly used with the goal to produce lower weight components with lower friction between them and to produce newer materials with improved thermal characteristics. Therefore, mechanical properties of Al-Si alloys have also been extensively investigated. By contrast, the corrosion resistance of Al-Si alloys is not sufficiently researched [18-25].

Silicon, as a major alloying element in aluminium alloys, is primarily responsible for good castability, low density (2.34 g cm^{-3}), and reduction of thermal expansion coefficient. Silicon is nobler than aluminium solid-solution matrix by several hundred mV and behaves like cathode in the matrix. However, the effects of silicon on the corrosion resistance of Al-Si based casting alloys are minimal because of low corrosion current density resulting from the fact that the silicon particles are highly polarized [6, 25].

Two important alloying elements that have been widely added to Al-Si casting alloys are copper and magnesium, which increase the strength of the alloy at room and elevated temperature and make them suitable for heat treatment. Copper-containing Al-Si alloys generally have lower corrosion resistance because copper can disperse in the aluminium oxide film and prevent complete passivation. The inhomogeneous distribution of copper in the alloy microstructure is a major cause of low resistance to pitting and stress corrosion cracking [26]. Magnesium provides improvement of the work hardening characteristics and can impart good corrosion resistance and weldability of alloys [5].

The presence of iron can decrease good alloy properties as this element represents the major class of intermetallic phases. Phases of Al-Fe-Si-Mg, which crystallize in the form of so-called "Chinese Script", are iron-magnesium phases that behave in a cathodic manner regarding the aluminium matrix. Investigations have been showed that content of iron higher than 0.4 % in Al-Si alloys could lead to an evaluation of sponge-like interdendritic porosity [5, 17, 25].

The increase of zinc content in the Al-Si casting alloys is often related to the aluminium alloys casting production from the secondary ingots that have been produced from the return aluminium scrap metals during recycling. Investigations of Saito and associates [27] have shown that increase in zinc content leads to modest increase in alloy hardness due to increased number densities of needle-shaped precipitates in the Al-Mg-Si alloy. Zinc atoms were incorporated in the precipitate structures at different atomic sites. Also, zinc atoms segregate along grain boundaries and lead to high intergranular corrosion susceptibility of alloy when zinc concertation is ≈ 1wt% [27]. Investigations of Morinaga and associates [28] of zinc effects on the properties of Al-Si-Cu die-casting alloys have shown small increase of strength associated with increase in zinc content in alloy and ~~also~~ increase in weight loss after performing saltwater immersion test which confirmed negative effect of zinc on corrosion resistance of alloy.

Adding manganese to aluminium alloys enhances tensile strength, improves low-cycle fatigue resistance and corrosion resistance as well [5].

In this paper, the results of electrochemical testing and microstructural characterization of Al-Si casting alloys have been shown and analyzed to investigate their corrosion properties. While previous researches were mostly based on results from testing samples prepared specifically for those researches, in this paper all experiments have been done on specimens made from finished parts and alloys used in automotive industry, manufactured at *Cimos* in Roč and Buzet, Croatia. Cimos factory in Buzet is using

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AlSi9Cu3 and AlSi12Cu1 aluminium alloys for manufacturing parts by high pressure die casting process, while factory in Roč is using AlSi12Cu1, AlSi9Cu3, AlSi7Mg and AlSi8Cu3 aluminium alloys for manufacturing parts by gravity die casting.

2 Experimental investigation

2.1 Materials

Investigated samples were prepared by cutting the automotive parts onto smaller pieces as shown in Fig. 1.
The chemical composition of investigated aluminium alloys is shown in Table 1. Abbreviations in the table refer to the following casting processes: HPDC - high pressure die casting, and GDC - gravity die casting. Glow discharge optical emission spectrometer LECO GDS 500 A has been used to determine the chemical composition of alloys.

Figure 1. Preparation of samples for microstructural and electrochemical investigations from the automotive parts.

Alloys in Table 1 are aluminium casting alloys of the 40000 series, based on the binary aluminium-silicon system and containing from 7 % up to 12 % of silicon that improves castability of aluminium. Maximum castability is achieved with eutectic composition of 13 % silicon.

2.2 Microstructural characterization

Microstructural characterization after casting has been done to obtain the connection between the microstructure and the corrosion resistance of investigated alloys. Metallographic samples have been prepared by grinding with Struers MD-Piano 220 resin bonded diamond disc, followed by MD-Allegro composite disc with diamond suspension.

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Table 1. Chemical composition of aluminium alloys (balance Al)

Sample no.	Alloy		Chemical composition [%]							Casting process
			Si	Fe	Cu	Mn	Mg	Zn	Cr+Ni+Ti	
1	AlSi9Cu3	46000	9.47	0.68	3.03	0.20	0.11	0.88	0.11	HPDC
2	AlSi12Cu1	47100	11.11	0.65	0.80	0.26	0.04	0.43	0.09	GDC
3	AlSi12Cu1	47100	10.69	0.63	0.71	0.26	0.09	0.44	0.10	HPDC
4	AlSi9Cu3	46000	8.61	0.67	2.91	0.20	0.12	0.81	0.09	GDC
6	AlSi7Mg	42000	7.10	0.11	0.10	0.07	0.30	0.01	0.14	GDC
7	AlSi8Cu3	46200	8.68	0.65	2.52	0.30	0.29	0.57	0.21	GDC

After grinding, samples have been polished with MD-Dac polishing cloth with DiaPro Dac 3 diamond suspension, followed by MD-Chem polishing cloth with OP-U NonDry colloidal silica suspension. After grinding and polishing, samples have been etched using “Keller’s etch” (32 % HCl, 66 % HNO_3, 40 % HF). An OLYMPUS BX51 optical microscope has

been used to observe microstructure from etched surfaces.
After electrochemical investigations, corroded sample surfaces have been rinsed by mixing in demineralized water and dried in desiccator and then examined with an optical microscope.

2.3 Electrochemical investigations

Electrochemical investigations have been performed using a Princeton Applied Research potentiostat Parstat 2263. A three-electrode set-up has been used: the working electrode with an exposed area of 1 cm^2, a saturated calomel electrode (SCE) placed in Luggin capillary as the reference electrode and graphite counter electrode.
Prior to electrochemical investigations, samples have been ground and polished with the sandpapers P320, P600, P800, P1200 and P2400, afterward degreased in ethanol and rinsed with deionized water.
All measurements were conducted at room temperature (20 ± 2 °C), in the 0.6 mol dm^{-3} NaCl naturally aerated solution, prepared from analytical grade NaCl and deionized water.
Open circuit potential, E_{OC}, measurements have been performed as a function of time for a period of 2 h. Potentiodynamic polarization has been performed at a scan rate of 0.166 mVs^{-1}, over a potential range of E_{OC} ± 250 mV, starting from the most negative potential.
The corrosion rate has been calculated from polarization curves, based on the corrosion current density, i_{corr}, known equivalent weight of the material, EW, and material density, ρ, (Table 2), according to the expression:

$$v_{corr} = \frac{0.0033\, i_{corr} \cdot EW}{\rho} [\text{mmpy}]. \qquad (1)$$

3 Results and discussion

The importance of microstructure to the properties of metals and alloys has long been recognized [29]. It has been established that the presence of alloying elements in the microstructure, such as insoluble intermetallic particles or single elements (copper, silicon), leads to the formation of local electrochemical cells between them and the aluminium matrix. These electrochemical cells can cause severe and highly localized attack by pitting corrosion in aggressive medium. The electrochemical nature of the intermetallic particles plays an important role in susceptibility of an aluminium alloy to localized corrosion [13, 30]. Therefore, at the beginning of the investigations, microstructure characterizations have been done and the results of these investigations are presented in Fig. 2.
Fig. 2 shows the microstructure of etched samples, where major micro-constituents can be seen: the primary α_{Al} phase and the eutectic (α_{Al} + β_{Si}) phase. Size and distribution of α_{Al} phase, as well as morphology and distribution of the eutectic (α_{Al} + β_{Si}) phase can be explained by the difference in solidification speeds between the surface and the center of the part [31-33].
The time dependence of the open circuit potential for samples of investigated aluminium alloys is shown in Fig. 3

Table 2. Density and equivalent mass of aluminium alloys

Sample no.	1	2	3	4	6	7
ρ [g cm^{-3}]	2.76	2.65	2.65	2.76	2.68	2.76
EW [g]	9.825	9.156	9.145	9.800	9.920	9.670

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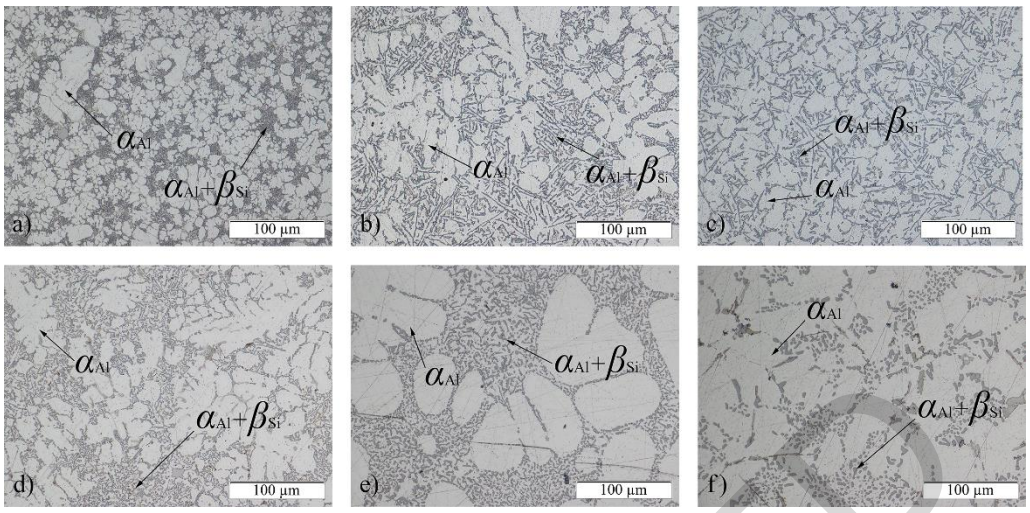

Figure 2. Microstructure of aluminium alloys after casting; sample no.: a) 1, b) 2, c) 3, d) 4, e) 6, f) 7.

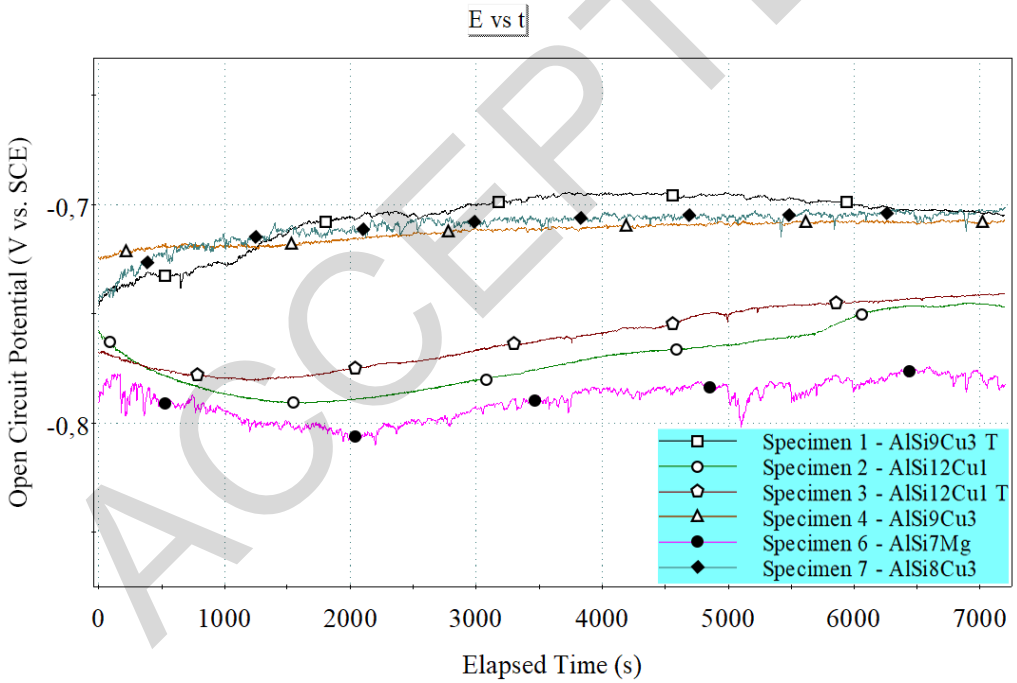

Figure 3. Open circuit potential vs. time curves for samples in 0.6 M NaCl solution.

The change of the open circuit potential, E_{OC}, is the consequence of the changes that occur due to the chemical reaction at the electrode/solution interface. Specifically, the established E_{OC} value depends on the conditions of the measurement being performed, the composition and state of the surface of the examined metal sample, the characteristics of the electrolyte solution, and the time of exposure of the metal sample to the electrolyte.

It can be seen that the initial values of E_{OC} for samples no.1 (AlSi9Cu3, HPDC), no. 4 (AlSi9Cu3, GDC) and no. 7 (AlSi8Cu3, GDC), which have a higher copper content, have been more positive than for other three alloy samples. Also, for above mentioned samples, E_{OC} values move in the positive direction immediately after immersion into the electrolyte solution, which can be attributed to the spontaneous formation of the corrosion product film at the electrode surfaces. The presence of copper in aluminium alloys raises the open circuit potential towards positive direction, which has been reported in literature [34, 35]. In case of sample no. 2 (AlSi12Cu1, GDC) and sample no. 3 (AlSi12Cu1, HPDC), initial drop of E_{OC} values can be attributed to

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the adsorption of negative chloride ions onto the electrode surfaces covered with an oxide film with slightly worse protective properties [35]. After 25 minutes the opposite trend can be observed, which indicates build-up of protective oxide film on electrode surface. For sample no. 6 (AlSi7Mg, GDC), E_{OC} values move in a positive direction after more than 30 minutes, with the slightest tendency to shift the curve in a positive direction.

Two phenomena related to integrity of air-formed oxide on the surfaces of the different alloys can be observed. The air-formed oxides appear to be stable in the 0.6 M NaCl electrolyte and do not undergo dissolution. Spontaneous positive displacements of E_{OC}, corresponding to the formation of corrosion product layer oxide film on the top of the air-formed oxide, have been observed.

On the other hand, specimens no. 2, no. 3, and no. 6, with lower Cu content, have negative displacement of E_{OC}, which corresponds to initial dissolution of the air-formed oxide in 0.6 M NaCl electrolyte. Positive displacement of the E_{OC} appears after 30 minutes.

Based on Fig. 3 it can be concluded that the type of casting process, HPDC or GDC, does not affect the E_{OC} values of investigated aluminium alloys. Content of Cu in the alloy has the greatest influence on E_{OC} values.

Potentiodynamic polarization curves, presented in the semi-logarithmic plot (Fig. 4), are composed of cathodic and anodic branches, which result from the electrochemical reactions in the system. The anodic branch of the polarization curve describes the anodic dissolution of aluminium in a NaCl solution, which can be expressed by equation:

$$Al \rightarrow Al^{3+} + 3e^{-} . \quad (2)$$

The cathodic part of the polarization curve represents an oxygen reduction reaction:

$$O_2 + 2H_2O + 4e^{-} \rightarrow 4OH^{-} . \quad (3)$$

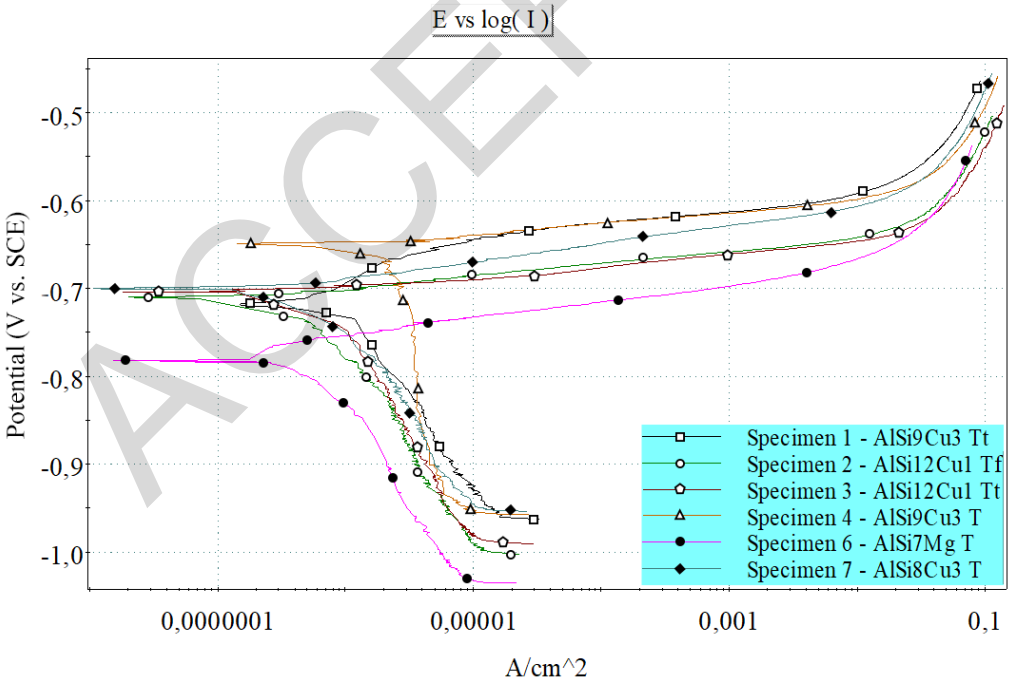

Figure 4. Potentiodynamic polarization curves for samples in 0.6 M NaCl solution.

Unlike pure aluminium [36], for all samples, the anodic current density increases rapidly with the potential indicating an active dissolution. The behavior observed in these curves is similar to that reported by Guillaumin et al. [37] and Rendon and Calderon [38]. It can be seen that the values of anodic current, i_a, are much higher than the cathodic current, i_c, at high overpotential values. This indicates that the reduction reaction occurs at slower rate than oxidation reaction, and therefore, cathodic reaction controls the electrochemical corrosion of aluminium alloy. From Fig. 4 it can be seen that the lowest values of cathodic current density were obtained for specimens no. 6, no. 2, and no. 3 which also have the

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highest values of anodic current densities. Opposite behavior can be seen with alloys no. 1, no. 4, and no. 7, which showed the lower values of anodic current density and the higher values of cathodic current density.

Corrosion parameters obtained from polarization measurements are shown in Table 3.

From Table 3 it can be seen that the four investigated alloys (samples no. 6, 2, 1 and 7) have quite low corrosion current density (i_{corr} < 0.4 μA cm^{-2}) which indicates their excellent corrosion resistance. The highest corrosion current density was determined for the sample no. 4 due to its highest cathodic current density. This alloy has high percentage of zinc in its composition, which could be the reason for this behavior.

Since this research is based on the investigations of alloys from finished parts, in which the content of all alloying elements varies, the conclusions about the influence of a particular alloying element on corrosion properties of investigated alloys are very difficult to present.

Fig. 5 shows the corrosion morphology of investigated samples after electrochemical testing. After polarization measurements, all investigated alloys have rough surfaces due to alloy dissolution processes. Round spots with different colors are randomly distributed on the surface of samples no. 1 to no. 4 (Fig. 5a to 5d) as indication of places on which the intensive electrochemical processes take place as a result of different composition of alloys and their inhomogeneity, which have been identified in different Cu-Si casting alloys in similar investigations [17, 18, 23, 25].

Table 3. Corrosion parameters of the samples taken from potentiodynamic polarization curves

Sample no.	E_{corr} [mV][1]	i_{corr} [μA cm^{-2}]	β_c [mV dec^{-1}][2]	β_a [mV dec^{-1}][3]	v_{corr} [mm py]
1	-686.1	0.310	-151.5	26.7	0.00361
2	-710.4	0.308	-177.1	15.4	0.00348
3	-701.5	0.882	-280.8	13.2	0.00995
4	-652.8	2.470	-823.8	16.4	0.02860
6	-772.6	0.174	-144.3	21.1	0.00190
7	-703.2	0.351	-159.7	22.1	0.00401

[1] Corrosion Potential; [2] Cathodic Tafel Slope; [3] Anodic Tafel Slope

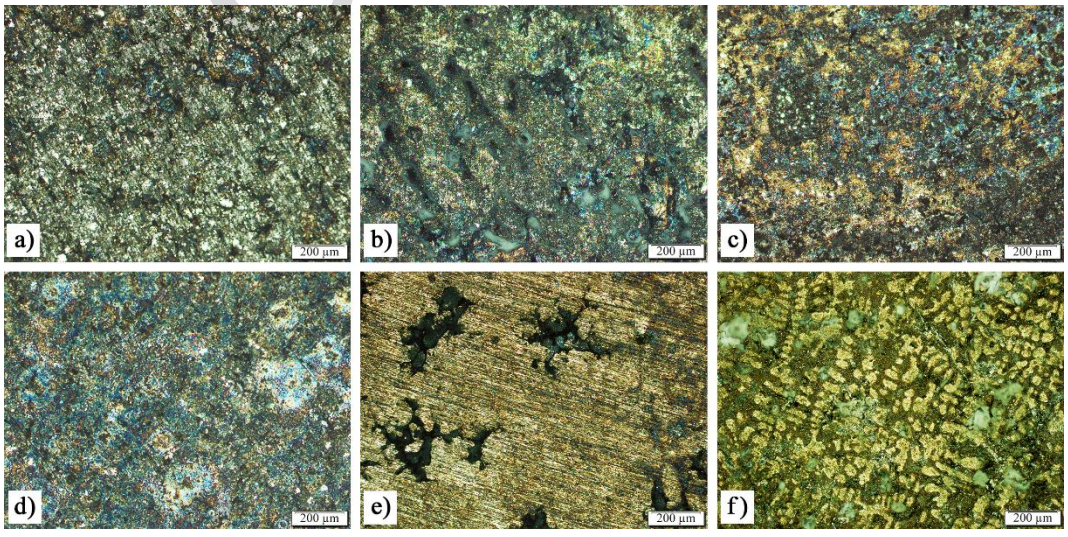

Figure 5. Corrosion morphology after electrochemical testing; sample no.: a) 1, b) 2, c) 3, d) 4, e) 6, f) 7.

One of the features which could have influence on the corrosion behavior of investigated alloys is related to the production technology of the casting alloy which could lead to the formation of some defects and

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impurities on the surface. These defects can become the nuclei of the corrosion attack on the casting parts. One of the most common defects of as-cast Al-Si alloys is the interdendritic porosity of the material, which causes the reduction of alloy resistance against corrosion [17, 25].

It is important to note that the values of the corrosion rates for all investigated alloys are sufficiently low, which confirmed that those alloys are suitable for manufacturing the automotive parts.

4 Conclusion

The corrosion behavior of six Al-Si alloys (40000 series), used in production of finished parts for automotive industry, has been analyzed by measuring open circuit potential, potentiodynamic polarization, as well as by optical microscopy examination.

Alloys with higher copper and silicon content have more positive values of open circuit potential.

All alloys have shown good corrosion resistance, which has manifested with the low values of corrosion rates calculated from the values of corrosion current densities from potentiodynamic polarization measurements.

Differences in electrochemical behavior appeared due to differences in their chemical composition and microstructure.

The alloying elements determine the overall corrosion behavior of as-cast Al-Si alloys.

The type of casting process does not affect the electrochemical behavior of investigated alloys.

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